

Amendments to the Claims:

This listing of claims will replace all prior versions and listings of claims in the application:

Listing of Claims:

Claim 1 (currently amended): An article of manufacture suitable for use in determining whether or in what amount a chemical species is present in a target environment, which article comprises a multiplicity of particles in three-dimensional close-packed orientation, said particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension, and on said core a ligand shell, of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with said a chemical species in a target environment such that a an electrical conductivity property of said multiplicity of particles is altered.

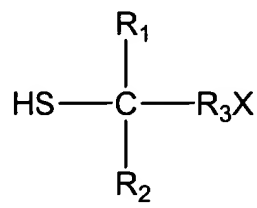
Claim 2 (original): An article of manufacture as defined in claim 1, wherein said core comprises silver, gold, platinum or palladium, or an alloy of two or more such metals.

Claim 3 (canceled)

Claim 4 (currently amended): An article of manufacture as defined in claim 1, wherein said ligand shell molecule comprises a thiol or an amine as the bonding site to the gold surface.

Claim 5 (currently amended): An article of manufacture as defined in claim 4, wherein said ligand shell comprises a thiol ligand molecule selected from the group consisting of primary aliphatic thiols, secondary aliphatic thiols, tertiary aliphatic thiols, heterofunctionally substituted

aliphatic thiols, ~~aromatic thiols, heterofunctionally substituted aromatic thiols,~~ and
heterofunctionally substituted araliphatic thiols as depicted by the general formula:



wherein for primary aliphatic thiols:

$\text{R}_1 = \text{R}_2 = \text{X} = \text{H}$; and

$\text{R}_3 = \text{C}_n\text{H}_{2n}$ ($n = 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17$, or 18);

for secondary aliphatic thiols:

$\text{R}_1 = \text{X} = \text{H}$;

$\text{R}_2 = \text{C}_{n'}\text{H}_{2n'+1}$ ($n' = 1, 2, 3, 4$, or 5); and

$\text{R}_3 = \text{C}_n\text{H}_{2n}$ ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15$, or 16);

for tertiary aliphatic thiols:

$\text{X} = \text{H}$;

$\text{R}_1 = \text{C}_{n''}\text{H}_{2n''+1}$ ($n'' = 1, 2, 3, 4$, or 5);

$\text{R}_2 = \text{C}_{n'}\text{H}_{2n'+1}$ ($n' = 1, 2, 3, 4$, or 5); and

$\text{R}_3 = \text{C}_n\text{H}_{2n}$ ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15$, or 16);

for heterofunctional aliphatic thiols:

$\text{R}_1 = \text{R}_2 = \text{H}$;

$\text{R}_3 = \text{C}_n\text{H}_{2n}$ ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17$, or 18); and

$\text{X} = \text{OH}, \text{F}, \text{Cl}, \text{Br}, \text{I}, \text{CF}_3, \text{CN}, \text{COOH}, \text{SO}_3\text{H}, \text{NO}_2, \text{NR}'\text{R}''$ ($\text{R}' = \text{H}, \text{CH}_3$, or C_2H_5

and $\text{R}'' = \text{H}, \text{CH}_3, \text{C}_n\text{H}_{2n+1}$ ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17$, or 18)), $\text{CO}_2\text{R}'$

($\text{R}' = \text{CH}_3, \text{C}_n\text{H}_{2n+1}$ ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17$, or 18)), OR' ($\text{R}' = \text{CH}_3$,

Applicant(s): Snow et al.

C_nH_{2n+1} ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17, \text{ or } 18$), $(CH_2CH_2O)_nH$ ($n = 1, 2, 3, \text{ or } 4$), $(CH_2CH_2O)_nCH_3$ ($n = 1, 2, 3, \text{ or } 4$), $CO_2NR'R''$ ($R' = H, CH_3 \text{ or } C_2H_5$; and $R'' = H, CH_3, C_nH_{2n+1}$ ($n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17, \text{ or } 18$)), $C(CF_3)_2OH$, piperidinyl, pyrazinyl, morphinyl, or imidazolyl

for heterofunctional araliphatic thiols:

$R_1 = R_2 = H$;

$R_3 = C_nH_{2n}C_6H_4$ ($n = 1, 2, 3, 4, 5, \text{ or } 6$); and

$X = OH, F, Cl, Br, I, CF_3, CN, COOH, SO_3H, NO_2, NR'R''$ ($R' = H, CH_3, \text{ or } C_2H_5$; and $R'' = H, CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)), CO_2R' ($R' = CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)), OR' ($R' = CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)), $(CH_2CH_2O)_nH$ ($n = 1, 2, 3, \text{ or } 4$), $(CH_2CH_2O)_nCH_3$ ($n = 1, 2, 3, \text{ or } 4$)), or $CO_2NR'R''$ ($R' = H, CH_3, C_2H_5$ and $R'' = H, CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)).

Claim 6 (original): An article of manufacture as defined in claim 4, wherein said ligand shell comprises an amine selected from the group consisting of primary aliphatic amines.

Claim 7 (original): An article of manufacture as defined in claim 1, wherein in each said particle the core is of size from 2 to 20 nm in maximum dimension and the ligand shell is of thickness from 0.4 to 2.5 nm.

Claim 8 (original): An article of manufacture as defined in claim 1, wherein the particles are substantially spherical.

Claim 9 (currently amended): An article of manufacture as defined in claim 1, wherein the ligand shell contains ligand molecules having a single bonding site to a gold surface and a second a heterofunctional group capable of ~~binding both with the core and~~ interacting with the analyte of interest.

Claims 10-20 (canceled)

Claim 21 (currently amended): An assembly suitable for investigation of a target environment to determine whether or in what amount a chemical species may be present, which comprises

- (a) a substrate suitably configured for presenting a multiplicity of particles supported thereon to contact with said environment;
- (b) supported by said substrate, a multiplicity of particles in three-dimensional close-packed orientation, said particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension; and having deposited thereon a ligand shell, of thickness from 0.4 to 4.0 nm; and composed of an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with ~~said~~ a chemical species in a target environment such that a an electrical conductivity property of said multiplicity of particles is altered; and
- (c) a sensor for monitoring said property of said multiplicity of particles.

Claim 22 (original): An assembly as defined in claim 21, wherein said core comprises silver, gold, platinum or palladium or an alloy of two or more of such metals.

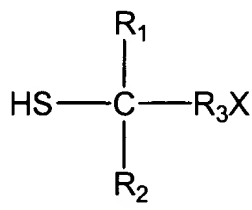
Claim 23 (canceled)

Claim 24 (currently amended): An assembly as defined in claim 21, wherein the film is of thickness from 5 to ~~40,000~~ 2,000 nm.

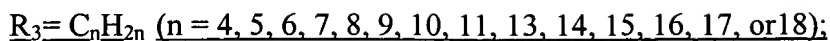
Claim 25 (currently amended): An assembly suitable for investigating a target environment, to determine whether or in what amount a chemical species may be present, which comprises

- (a) a substrate suitably configured for presenting a multiplicity of particles in three-dimensional close-packed orientation supported thereon to contact with said species;
- (b) supported by said substrate, said multiplicity of particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension, and ~~deposited thereon~~ having a ligand shell, of thickness from 0.4 to 4.0 nm; and composed of an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with said a chemical species in a target environment such that the electrical conductivity of particles is altered;
- (c) a pair of electrodes, each in electrical contact with said multiplicity of particles; and
- (d) a sensor for monitoring the electrical conductivity of said multiplicity of particles to determine whether there is, or the amount of, any change in said conductivity as an indication of whether or in what amount said species is present.

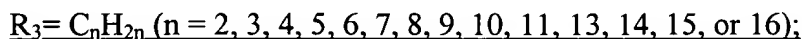
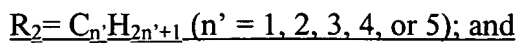
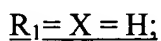
Claim 26 (currently amended): An assembly as defined in claim 25, wherein the core comprises gold and the ligand is selected from the group consisting of primary aliphatic thiols, secondary aliphatic thiols, tertiary aliphatic thiols, heterofunctionally substituted aliphatic thiols, ~~aromatic thiols, heterofunctionally substituted aromatic thiols,~~ and heterofunctionally substituted araliphatic thiols as depicted by the general formula:



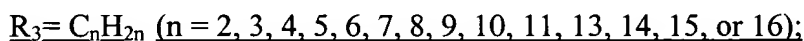
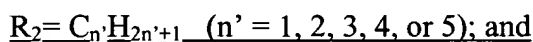
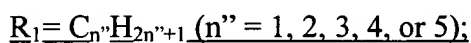
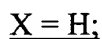
wherein for primary aliphatic thiols:



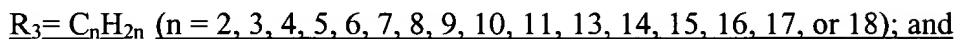
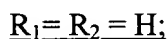
for secondary aliphatic thiols:



for tertiary aliphatic thiols:



for heterofunctional aliphatic thiols:



and R'' = H, CH}_3\text{, C}_n\text{H}_{2n+1} \text{ (n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17, or 18)), CO}_2\text{R'}

(R' = CH}_3\text{, C}_n\text{H}_{2n+1} \text{ (n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17, or 18)), OR' (R' = CH}_3\text{,}

C}_n\text{H}_{2n+1} \text{ (n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17, or 18), (CH}_2\text{CH}_2\text{O)}_n\text{H (n = 1, 2, 3, or}

4), (CH}_2\text{CH}_2\text{O)}_n\text{CH}_3 \text{ (n = 1, 2, 3, or 4)), CO}_2\text{NR'R'' (R' = H, CH}_3\text{, or C}_2\text{H}_5\text{; and R'' =}

H, CH}_3\text{, C}_n\text{H}_{2n+1} \text{ (n = 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 13, 14, 15, 16, 17, or 18)), piperidinyl, pyrazinyl,

morphinyl, or imidazolyl

for heterofunctional araliphatic thiols:



$R_3 = C_nH_{2n}C_6H_4$ ($n = 1, 2, 3, 4, 5, \text{ or } 6$); and

$X = OH, F, Cl, Br, I, CF_3, CN, COOH, SO_3H, NO_2, NR'R''$ ($R' = H, CH_3, \text{ or } C_2H_5$
and $R'' = H, CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)), CO_2R' ($R' = CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)), OR' ($R' = CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)), $(CH_2CH_2O)_nH$ ($n = 1, 2, 3, \text{ or } 4$), $(CH_2CH_2O)_nCH_3$ ($n = 1, 2, 3, \text{ or } 4$)), or $CO_2NR'R''$ ($R' = H, CH_3, C_2H_5$ and $R'' = H, CH_3, C_nH_{2n+1}$ ($n = 2 \text{ or } 3$)).

Claim 27 (currently amended): A method of fabricating an assembly suitable for investigation of a target environment to determine whether or in what amount a chemical species may be present, which comprises

(a) depositing on a substrate (i) a pair of interdigitated electrodes each having a comb-like configuration and (ii) in such manner that the electrodes are electrically connected, a thin film of a multiplicity of particles in a three-dimensional close-packed orientation having a core of conductive metal or conductive metal alloy, in each said particle the core being from 0.8 to 40.0 nm in maximum dimension, and ~~deposited on said core~~ having a ligand shell, of thickness from 0.4 to 4.0 nm; and composed of an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with said a chemical species in a target environment such that a an electrical conductivity property of said multiplicity of particles is altered; and

(b) connecting said pair of electrodes with a sensor capable of determining a change in the property of said multiplicity of particles.

Claim 28 (currently amended): A method of fabrication as defined in claim 27, wherein said deposition of a thin film of the multiplicity of particles comprises spraying on the electrodes and on the substrate a solution comprising ~~a salt of each conductive metal to be incorporated in said core, an organic substance having a functional group which is capable of interacting with~~

Applicant(s): Snow et al.

~~said species, and a solvent for each said salt and said substance~~ the multiplicity of particles and a solvent, said electrodes and substrate being at a temperature such that the solvent is flashed away or rapidly evaporated.

Claim 29 (currently amended): A method of fabrication as defined in claim 27, wherein said deposition of a thin film of the multiplicity of particles comprises

(a) treating said electrodes and substrate with a difunctional material capable of binding with (i) the electrodes and the substrate and (ii) said multiplicity of said particles, such that said material binds with said electrodes and said substrate;

(b) contacting the treated electrodes and substrate with said multiplicity of particles having a core of conductive metal or conductive metal alloy, in each said particle the core being from 0.8 to 40.0 nm in maximum dimension, and deposited on said core a ligand shell, ~~of having a thickness from 0.4 to 4.0 nm; and a composition as an encapsulating monomolecular layer of ligand molecules, each molecule having a single bonding site to a gold surface, which is the ligand shell being~~ capable of interacting with said species such that a an electrical conductivity property of said multiplicity of particles is altered, such that said multiplicity of particles bonds with the material to form a composite comprising a layer of said particles on the electrodes and substrate.

Claim 30 (original): A method of fabrication as defined in claim 29, wherein the multiplicity of particles forms a monolayer on said substrate and electrodes.

Claim 31 (currently amended): A method of fabrication as defined in claim 27, which further comprises a cycle of steps including

(a) exposing the outwardly facing surfaces of said particles of the composite to a coupling agent capable of bonding said particles to a further multiplicity of such particles

deposited thereon, the particles of said further multiplicity having a core of conductive metal or conductive metal alloy, in each said particle the core being from 0.8 to 40.0 nm in maximum dimension, and deposited on said core a ligand shell, ~~of having a~~ thickness from 0.4 to 4.0 nm, and of composition as an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with said species such that a an electrical conductivity property of said multiplicity of particles is altered; and

(b) contacting the particle surfaces so exposed with said further multiplicity of particles such that said further multiplicity of particles bonds with the particle surfaces of said composite, and the further multiplicity of particles is immobilized on those surfaces.

Claim 32 (original): A method of fabrication as defined in claim 31, wherein said cycle is performed a plurality of times.

Claim 33 (currently amended): A system suitable for investigating a target environment to determine whether or in what amount a chemical species may be present, which comprises

(a) a multiplicity of particles in three-dimensional close-packed orientation, said particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension, and ~~having deposited thereon~~ a ligand shell, of thickness from 0.4 to 4.0 nm, and composed of an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with said a chemical species in a target environment such that a an electrical conductivity property of said multiplicity of particles is altered;

(b) means for exposing said multiplicity of particles to said environment;

(c) means for subjecting said multiplicity of particles to conditions sufficient for said property to be exhibited; and

(d) means for monitoring said property to determine whether there is, or the amount of, any change in such property as an indication of whether or in what amount said species is present.

Claim 34 (currently amended): A system for investigating a target environment to determine whether or in what amount a chemical species may be present, which comprises

(a) a multiplicity of particles in three dimensional close-packed orientation, said particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension, and having deposited thereon a ligand shell; of thickness from 0.4 to 4.0 nm; and composed of an encapsulating monomolecular layer of ligand molecules each molecule having a single bonding site to a gold surface, which is the ligand shell being capable of interacting with said a chemical species in a target environment such that the electrical resistivity of said multiplicity of particles is altered;

(b) means for exposing said multiplicity of particles to said environment;

(c) means for passing an electrical current through said multiplicity of particles; and

(d) means for monitoring the electrical resistivity of said multiplicity of particles to determine whether there is, or the amount of, any change in said resistivity as an indication of whether or in what amount said species is present.

Claim 35 (original): A system as defined in claim 34, wherein said means for monitoring the electrical resistivity of said multiplicity of particles includes a current-to-voltage converter circuit followed by a precision rectifier and low-pass filter.

Claim 36 (original): A system as defined in claim 35, wherein said means further includes a voltage-to-frequency converter.

Claim 37 (new): An article of manufacture as defined in claim 5, wherein said heterofunctionally substituted aliphatic thiol or said heterofunctionally substituted araliphatic thiol is substituted by OH, COOH, NH₂, Cl, or combinations thereof.

Claim 38 (new): An article of manufacture as defined in claim 5, wherein said heterofunctionally substituted aliphatic thiol or said heterofunctionally substituted araliphatic thiol is substituted by HS(CH₂)₆OH or the hexafluoroacetone adduct.

Claim 39 (new): A method of investigating a target environment to determine whether or in what amount a chemical species may be present therein, which comprises:

(a) exposing to said environment an article of manufacture comprising a multiplicity of particles in close-packed orientation, said particles having a core of conductive metal or conductive metal alloy and deposited thereon a ligand which is capable of interacting with said species such that a property of said multiplicity of particles is altered wherein the ligand shell contains ligand molecules having a single bonding site to a gold surface and a second heterofunctional group capable of interacting with a chemical species in a target environment;

(b) subjecting said multiplicity of particles to conditions sufficient for said property to be exhibited; and

(c) monitoring said property to determine whether there is, or the amount of, any change as an indication of whether, or in what amount, said species is present.